Search for hidden orbital currents and observation of an activated ring of magnetic scattering in the heavy fermion superconductor URu_2Si_2

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(Received 3 October 2003; revised manuscript received 18 February 2004; published 30 April 2004)

We have performed neutron-scattering experiments on the heavy fermion superconductor URu_2Si_2 to search for the orbital currents predicted to exist in the ordered phase below $T_N=17.5$ K which result in a ring in momentum space. Elastic scans in the $(H, K, 0)$ and $(H, 0, L)$ planes revealed no such order parameter at low temperatures. This shows that any orbital current formation is quite small and less than our detection limit for a ring of scattering of $0.06(1)\mu_B$ (albeit somewhat greater than the size of the predicted moment of $\sim 0.03\mu_B$). On heating, however, we find that a ring of quasielastic scattering forms in the $(H, K, 0)$ plane centered at an incommensurate radius $\tau=0.4$ from the (1, 0, 0) antiferromagnetic (AF) Bragg position. The intensity at a point on the ring, (1.4, 0, 0), is thermally activated below T_N with a characteristic energy scale of $\Delta=110$ K$\sim 6T_N$. This is the coherence temperature, and it is much higher than the spin-wave energy for the selected momentum. We believe that the incommensurate spin fluctuations compete with the AF spin fluctuations, drive the transition to a disordered magnetic state above T_N, and contribute to the formation of the heavy fermion state.

DOI: 10.1103/PhysRevB.69.132418 PACS number(s): 75.25.+z, 74.70.Tx, 63.20.Dj

High-temperature superconductivity, superfluidity, the quantum Hall effect, and heavy fermion metals are all examples of the fascinating behavior that arises from complicated interactions within systems of fermions.[1,2] The interest in heavy fermion metals has been piqued by the discovery of the coexistence of superconductivity and antiferromagnetism in several species such as UPt_3,[3] UPd_2Al_3,[4] UNi_2Al_3,[5] and URu_2Si_2.[6,7] A striking feature of these compounds is that they display conduction-electron specific heats at low temperatures that are orders of magnitude greater than those found in typical metals. This behavior differs markedly from that at high temperatures where the value for the Sommerfeld constant returns to normal free-electron values. The resistivity, specific heat, and magnetic susceptibility show that these intermetallics behave at high temperatures as a set of weakly interacting conduction electrons and local moments. The crossover from this state to a low-temperature state, in which the effective mass of the quasiparticles increases dramatically, is a gradual change that is characterized by a coherence temperature θ_C.[8]

For UPt_3 and URu_2Si_2, the ordered moments in the Néel states are extremely small ($0.028\mu_B$[9] and $0.03\mu_B$,[6,7] respectively). For URu_2Si_2 the superconductivity below T_C ~ 1.2 K emerges from an unknown state with a transition at $T_N=17.5$ K. It is accompanied by a large λ anomaly in the specific heat,[10] which, in light of the extremely small ordered moment, suggests that another order parameter is at play.[11] There has been considerable recent interest in URu_2Si_2, with new developments providing hints that the ordered magnetic state is inhomogeneous,[12] and somewhat parasitic to a so-called ''hidden'' ordered state. A complex phase diagram for this state has been elucidated based upon specific-heat measurements at high magnetic fields.[13] The character of this possible hidden order parameter is still unknown, but possibilities such as quadrupolar ordering and charge-density wave formation are plausible.[14] It has been shown, however, that none of the allowed quadrupolar or octupolar orderings can account for the weak moment.[15,16] Recent NMR measurements, which have shown that small isotropic magnetic fields develop below T_N at the silicon sites,[17,18] have led Chandra *et al.* to propose that the hidden order arises from the formation of orbital currents.[19,20] The signature for such currents, which develop in the ordered phase below T_N, would be a ring of magnetic incommensurate scattering in reciprocal space with a characteristic Q^{-4} form factor.

We have carried out a detailed search for this hidden order parameter in URu_2Si_2 using neutron scattering in the $(H, K, 0)$ and $(H, 0, L)$ planes. We will show that any such scattering is small and lies below our detection limit. We will also show that a ring of quasielastic scattering exists that decreases in spectral weight below T_N.

Our experiments were performed at the DUALSPEC triple-axis spectrometer at Chalk River Laboratories with a focusing pyrolytic graphite (PG) monochromator and PG analyzer set to a fixed energy of 3.52 THz. A PG filter was used to remove higher-order contamination. Elastic scans were performed, as well as quasielastic scans at an energy transfer of 0.25 THz. The collimation was 0.40°-0.48°-0.56°-1.20°. The two crystals were described earlier,[6,7] one oriented in the $(H, K, 0)$ plane and the other in the $(H, 0, L)$ plane.

Figure 1 shows the (1, 0, 0) magnetic Bragg signal arising from the $0.03\mu_B$ ordered moment. The relative intensity of this peak as compared to background gives a measure of our

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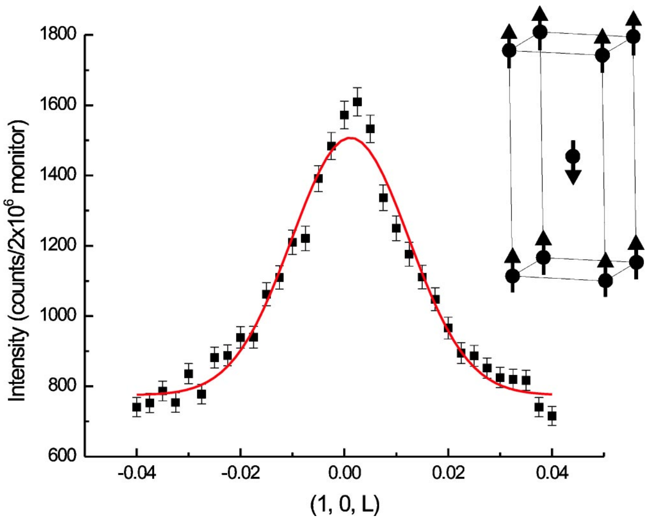

FIG. 1. The antiferromagnetic Bragg peak at (1, 0, 0) and $T=8$ K (fit is to a Gaussian). The inset shows the corresponding magnetic structure for the U^{4+} moments.

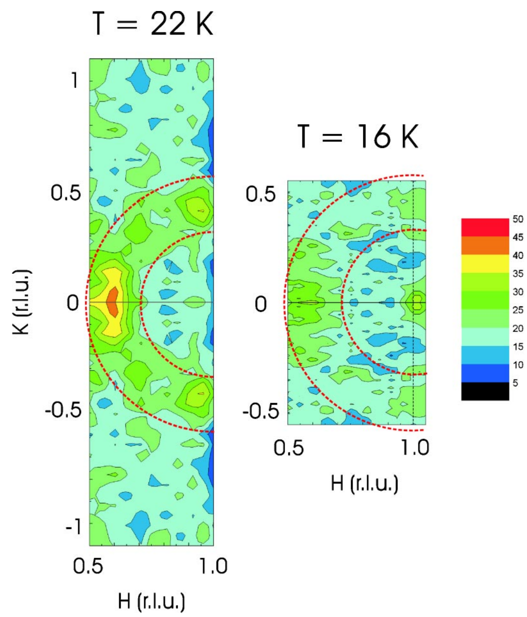

FIG. 2. (Color online) Contour plot of scattering in the $(H, K, 0)$ plane at $T=22$ K and $T=16$ K with $\Delta E=0.25$ THz energy transfer. Note that the scattering lies within a ring of momenta centered on (1, 0, 0), and the antiferromagnetic fluctuations below T_N at (1, 0, 0) associated with the order parameter. The dashed circles are guides to the eye.

sensitivity to small moments. From the ability to detect signal above background, we calculate that the minimum detectable signal for long-ranged local-moment formation is $0.013(1)\mu_B$ for three-dimensional (3D) order. For a moment distribution which leads to a 2D ring structure with the 0.2 r.l.u. (reciprocal lattice units) radius of Chandra *et al.*,[19] this limit changes to $0.06(1)\mu_B$. We calculated the reduced sensitivity by distributing the detectable 3D moment squared $(0.013\mu_B)^2$ located in an area $\pi(\Delta q_{coll}/2)^2$, over a ring of predicted radius $\tau=0.2$ r.l.u.[19] The factor by which the sensitivity is reduced is then

$$\frac{2\pi\tau\Delta q_{coll}}{\pi(\Delta q_{coll}/2)^2}\sim 24, \tag{1}$$

where Δq_{coll} is the instrumental resolution (in Å^{-1}). Thus the moment sensitivity is reduced to $0.013\mu_B\sqrt{24}=0.06\mu_B$. We note that Bull *et al.* quote only their detection limit $0.007\mu_B$ for a 3D peak, which is not appropriate for a ring of 2D scattering.[21]

The hidden order search was made with elastic raster scans with ranges $0.5\leqslant H\leqslant 1.0$ and $0\leqslant K\leqslant 1.05$ over the $(H, K, 0)$ plane and $0.175\leqslant H\leqslant 1.075$ and $0\leqslant L\leqslant 1.05$ over the $(H, 0, L)$ plane (with a step size of 0.025 r.l.u.). Note that for body-centered tetragonal symmetry, the position for the predicted ring of scattering at $(\tau\cos\theta,\ \tau\sin\theta,\ 1)$ (Refs. 19 and 20) is equivalent to the wave vectors $(1+\tau\cos\theta,\ \tau\sin\theta,\ 0)$ at which we made the search, albeit at different magnitudes of Q. In the difference scans, $I(8\text{ K})-I(22\text{ K})$, we found no additional signal from magnetic Bragg or ring scattering below T_N. This indicates that no new magnetic Bragg peaks are observable in the $(H, K, 0)$ and $(H, 0, L)$ planes. Bull reached the same conclusion for the (H, H, L) plane.[21] However, this does not completely rule out orbital current formation, for our detection limit for a ring of scattering, $0.06(1)\mu_B$, exceeds the predicted moment of Chandra of $\sim 0.03\mu_B$.[19,20] The rapid $1/Q^4$ form factor decrease can also render detection difficult.

Since no new features were discovered in the elastic channel, we decided to look at the quasielastic spectra at $\Delta E=0.25$ THz (just outside our energy resolution). This removes the large incoherent elastic peak and so increases the sensitivity to the formation of slow correlations modulated in **Q**. Our strategy was to further investigate an incommensurate ring of scattering which was discovered in the previous investigation of the inelastic spectrum near (1.4, 0, 0). A broad feature centered at about 0.6 THz was reported above T_N, indicative of heavily damped antiferromagnetic (AF) spin fluctuations.[6,7] This feature sharpened to a resolution limited peak with energy 1.1 THz, well below T_N with a center at higher energies ($\sim$1.1 THz). The increase in the incommensurate scattering as one passes above T_N (Ref. 22) originates from the downward shift and increased damping of the spin fluctuations (the location of a minimum in the spin-wave dispersion). The structure of this scattering in reciprocal space at this energy transfer was not reported in the original paper, nor was its explicit temperature dependence.[6] Unpublished work suggested that the structure could be a ring in reciprocal space,[23] a structure reminiscent of the ring that Chandra *et al.* predicted for orbital current formation in the hidden order phase.

Figure 2 shows contours of the scattering in the $(H, K, 0)$ plane at 22 K and 16 K, above and below T_N, respectively. We have folded the data about the line $K=0$ because of the symmetry of reciprocal space. The ringlike modulation comes from antiferromagnetic spin fluctuations [since we find it centered on other AF points such as (2, 1, 0), but not on the ferromagnetic point (1, 1, 0), as included

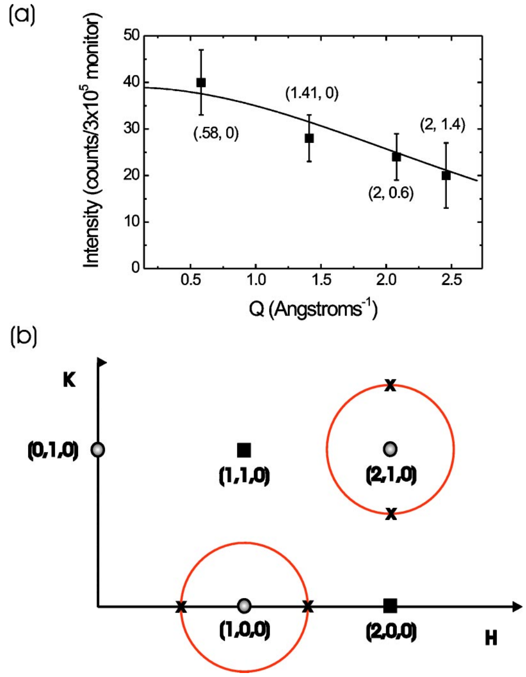

FIG. 3. (Color online) (a) The background subtracted intensities (as determined by Gaussian fits) to the features seen at rings of scattering in the (H, K, 0) plane at $T = 22$ K. The line is the U^{4+} magnetic form factor (Ref. 24). (b) A map of the rings in reciprocal space with crosses where the form factor was measured. The squares and circles refer to ferromagnetic and antiferromagnetic points, respectively.

on the mesh scan]. To confirm the modulation is magnetic in origin, we measured the form factor out to higher values of **Q**. Figure 3 shows the peak intensities at (0.6, 0, 0), (1.4, 0, 0), (2, 0.6, 0), and (2, 1.4, 0) derived from raster scans. The U^{4+} magnetic form factor in the same figure is in good agreement with our data, showing that the modulation arises from $5f$-shell electrons and not from larger diameter orbital currents. The latter would have exhibited the rapid Q^{-4} decrease Chandra *et al.* predicted for orbital currents.

The intensity on the ring of scattering sampled at (1.4, 0, 0) is thermally activated up to the transition at T_N as shown

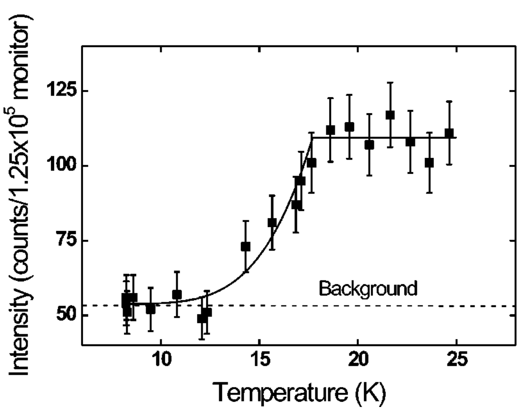

FIG. 4. The temperature dependence of the scattering at (1.4, 0, 0) and $\Delta E = 0.25$ THz energy transfer. The fit is with an activation temperature of 110(10) K.

in Fig. 4. There is some intensity modulation around the ring at 22 K, even when the 16 K data are subtracted, and it favors fluctuations in the (1, 0, 0) and (0, 1, 0) directions. The suppression along intermediate directions is likely because it takes more energy to create a spin fluctuation along the (1, 1, 0) direction, relative to (1, 0, 0) for the same radial momentum about (1, 0, 0).[7] The 22 K intensity at (0.6, 0, 0) exceeds that at (1.4, 0, 0) by more than the form-factor difference and is not understood. Even where there is the least scattering on the ring, the intensity there declines on entering the ordered phase whereas the orbital current signal should increase. Despite the unexplained modulation of the ring the intensity everywhere is consistent with activated behavior as shown in Fig. 4. Placed in context of the work of Broholm *et al.*,[6,7] who measured inelastic spectra above and below the transition, what we are observing above T_N is the tail of a highly damped, almost quasielastic spin fluctuation. Below T_N the scattering moves above our 0.25 THz energy window as a spin wave develops, narrows, and moves to a higher energy of ~ 1.1 THz, resulting in a suppressed intensity at 0.25 THz.

The fit in Fig. 4 is to a background plus a single activated intensity of the form

$$I(T) = A\exp(-\Delta/T), \quad T < T_N, \tag{2}$$

$$I(T) = \text{const}, \quad T > T_N, \tag{3}$$

where A is a constant, T_N is the Néel temperature (17.5 K), and $\Delta = 110(10)$ K is the fitted activation energy. An important result is that the activation temperature is not that of the sampling energy, 0.25 THz$\sim$12 K, nor that of the 0.6 THz $\sim$30 K spin excitation above T_N, nor that of 1.1 THz $\sim$53 K excitation below T_N. Instead it is the much larger activation energy found by Palstra *et al.*[10] for the specific-heat anomaly below T_N, who extracted it from a fit over the range 2 K–17.5 K to

$$C(T) = \gamma T + \beta T^3 + \delta\exp(-\Delta/T), \tag{4}$$

and found $\Delta \sim 115$ K. This suggested that a substantial gap opens in the density of states below T_N that effectively removes $\sim$75% of the low-lying states from the Fermi surface.[10] The gap energy is similar to the maximum in the spin-wave density of states,[25] and to the charge gap seen in the optical reflectance measurements.[26] Taken with our results, this serves to confirm that removal of both low-energy spin states and charge states is required to form the unknown ordered state below T_N. The existence of two competing energy scales (Δ and T_N) has also been observed in dc resistivity experiments.[27]

The quasielastic scattering observed in our experiment is reminiscent of the intimate connection between spin fluctuations and the heavy fermion state. Gaulin *et al.* studied this relationship in UNi_2Al_3, which has a T_N of 4.6 K and T_c of 1.2 K.[28,29] The magnetic structure is incommensurate, with an ordering wave vector of $\mathbf{Q} = (0.5 \pm \tau, 0, 0.5)$ ($\tau = 0.11$) and an ordered moment of $0.85\mu_B$ per U atom. The quasielastic spin fluctuations are of two kinds: those associated with the incommensurate wave vector and with the commensurate $\mathbf{Q}=(0, 0, 0.5)$ wave vector. The two modes compete with one another, with a shift in spectral weight from the commensurate to incommensurate fluctuations below T_N. Above the transition, the incommensurate fluctuations disappear, but the commensurate ones persist to nearly 80 K. Since this is the coherence temperature, the excitations are therefore associated with the formation of the heavy fermion state.

For URu_2Si_2, the situation is reversed: the commensurate fluctuations are associated with the ordering wave vector (1, 0, 0), and the incommensurate excitations persist to high temperatures,[22] and so can be identified with the formation of the heavy fermion state. This shift in spectral weight can be noted by the increase in scattering at (1, 0, 0) below T_N (see Fig. 2) and the corresponding decrease in intensity at (1.4, 0, 0). Mason *et al.* have suggested that magnetic frustration plays a role in the unusual magnetic properties of URu_2Si_2, with the long-range oscillatory nature of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction providing the mechanism.[22] The incommensurate ring of scattering we observe may be a signature of such a RKKY interaction. However, the nature of the fluctuations cannot be ascertained with this experiment alone. It is already known[7] that the spin fluctuations carry a matrix element of $1.2\mu_B$, two orders of magnitude larger than the weak moment of $0.03\mu_B$. Based on our experimental results that the activation energy is the same for the spin response and the specific-heat anomaly, we suggest that these are excitations out of the gapped "hidden order" state rather than the weakly AF ordered phase. Further experiments are needed to develop a clearer picture of the origin of these excitations with respect to proposed scenarios of a phase separation occurring at T_N. It may be that this feature is linked to a possible electronic phase separation, as suggested by muon-spin-resonance[12] and NMR measurements,[17] since the spectral weight is too large to be explained by the $0.03\mu_B$ ordered moment.

In conclusion, our neutron-scattering measurements in search of hidden order in the $(H, K, 0)$ and $(H, 0, L)$ planes have placed an upper limit of $0.013(1)\mu_B$ for the presence of any long-ranged 3D ordered spin structure well defined in **Q**. For a ring of scattering, the detectable moment is $0.06(1)\mu_B$, which precludes orbital current formation down to a level that is somewhat larger than that predicted by Chandra *et al.*.[19] Quasielastic scattering experiments have revealed a connection between a ring of incommensurate scattering at a radius of $\tau=0.4$ from the zone center and the heavy fermion state. The exponential activation energy of this ring below T_N, comparable with the specific-heat activation energy, suggests that a gap of 110 K is a feature of the hidden order phase.

C.R.W. acknowledges support from NSERC. The authors are grateful for the financial support of NSERC and CIAR. We thank B. D. Gaulin and C. Broholm for useful discussions and P. Chandra, P. Coleman, J. A. Mydosh, and V. Tripathi for a critical reading of this manuscript. The NRC technical staff at Chalk River, A. Cull, R. Sammon, and M. Potter, provided excellent support.

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